



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of Tetsuji Mitsumoto et al.

Serial No.: 09/928,093

5 Filed: April 6, 2001

Title: METHOD FOR PRODUCTION OF EASILY POLYMERIZABLE  
SUBSTANCE AND PURIFICATION APPARATUS

Art Unit: 1764

Examiner: MANOHARAN, Virginia

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COMMISSIONER FOR PATENTS

WASHINGTON, D. C. 20231

DECLARATION UNDER 37 C. F. R. 1.132

15 Sir:

I. I, Tetsuji Mitsumoto, of 241-1-203, Ebisu, Shikama-ku,  
Himeji-shi, Hyogo 672-8057, Japan, hereby declare that I am  
one of the inventors named in the above-mentioned application,  
20 that I graduated from Kobe University, Department of  
Engineering and Chemical Engineering in March 1991 with a  
B.S. degree. Since April 1991, I have been employed by Nippon  
Shokubai Co., Ltd., and engaged in chemical engineering  
research in the company from 1997.

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II. I understand that the above application has been  
rejected under 35 U. S. C. 103 (a) as being obvious over  
Matsumoto et al (U.S. 6,214,174) with or without Nutter (US  
4,304,738) and I have reviewed these references. It is of  
30 my opinion that the claimed methods in the present application  
are nonobvious over the cited references because the claimed  
methods achieved superior results as shown by the following

experimental results.

#### EXPERIMENT 1 (Example 1 of the present application)

A distillation column measuring 1400 mm in diameter and  
5 equipped therein with 10 dualflow trays having the opening  
area ratio of 23%, respectively, was provided in the lower  
part thereof with two dualflow trays of opening area ratio  
of 40% as splash collision plates. Using the distillation  
column, the purification of acrylic acid was done under the  
10 conditions of 4000 kg/h of feed quantity (purity of acrylic  
acid: 95% by weight, supplied from the middle part of the  
column), 2500 kg/h of distillation quantity (distilled from  
the top part of the column), 2500 kg/h of reflux quantity  
(extracted from the top part of the column and supplied from  
15 the top part of the column), 1 kg/h of feed rate of hydroquinone  
monomethyl ether as a polymerization inhibitor to the reflux  
liquid, 10 kg/h of feed amount of oxygen to the bottom of  
the distillation column, 6.0 kPa abs of column top pressure,  
and 70°C of column top temperature.

20 The dualflow tray was fixed, as illustrated in Fig. 5,  
by opening an oblong opening 20 x 40 mm in the tray and in  
the support ring welded to the wall of the column and fastening  
with a M10 (JIS) hexagonal bolt, a special square washer and  
special square nut measuring 50 x 20 mm, respectively, to  
25 the support ring and the tray in such a manner that the longer  
sides of the oblong opening and the longer sides of the washer  
and nut formed a roughly right angle. The number of the oblong  
opening was 20. 20 Segmental circular openings measuring 20  
mm in radius were provided on the welding line between the  
30 support ring and the wall of the column. The apparatus thus  
constructed was continuously operated for two months and then  
brought to a stop. When the interior of the apparatus was

visually inspected, the inner wall of the column, the upper sides of the trays, and the tray-fixing parts showed no sign of adhesion of a polymer. Though dirt originating in a polymer was observed on the rear side of the lowermost dualflow tray  
5 having an opening area ratio of 40%, abnormality such as blockage of openings, which would affect the operation of the distillation column, was not detected.

#### EXPERIMENT 2 (Example 2 of the present application)

10 The purification of acrylic acid was performed by using the same apparatus and the same conditions as in Example 1 while using vertical clamps, as illustrated in Fig. 6, in fixing the trays to the support rings.

The apparatus was continuously operated for two months  
15 and then brought to a stop. When the interior of the apparatus was visually inspected, the adhesion of a polymer to the gaps between the vertical clamps and the trays was observed and the polymer apparently fallen from the clamps accumulated on the outer peripheral parts of the trays underlying the  
20 clamps. The amount of the polymer so accumulated was about 3 kg per stage. The operating conditions were not confused during the two months' continuous operation in spite of this accumulation of the polymer.

#### 25 COMPARATIVE EXPERIMENT 1 (Comparative Example 1 of the present application)

The purification of acrylic acid was performed by using the same apparatus and the same conditions as in Example 1 while using horizontal clamps, as illustrated in Fig. 1, in  
30 fixing the trays to the support rings.

The apparatus was set operating as aimed at two months' continuous operation. From the start of the operation, the

pressure in the lower part of the distillation column was observed to rise with the elapse of time. Thus, the apparatus was stopped after one month's operation. When the interior thereof was visually inspected, the adhesion of a polymer to the gaps between the horizontal clamps and the trays was observed and the polymer apparently fallen from the clamps accumulated on the outer peripheral parts of the trays underlying the clamps. The amount of the polymer so accumulated was about 50 kg per stage. The accumulated polymer was clearly observed to initiate further advance of polymerization on the trays.

#### COMPARATIVE EXPERIMENT 2 (Comparative Example 2 of the present application)

The purification of acrylic acid was performed by using the same apparatus and the same conditions as in Example 1 while eliminating the liquid passing openings on the welding line between the support ring and the wall of the column, and fixing the trays as illustrated in Fig. 11. Fig. 11 is a diagram depicting the manner of fixing trays; Fig. 11A representing a partial cross section, Fig. 11B a diagram illustrating Fig. 11A as viewed from above (the direction of the space embracing bolts), and Fig. 11C a view illustrating Fig. 11A as viewed from below (the direction of the space embracing nuts). In Fig. 11, openings 11 mm in inside diameter were opened as bolt openings in a support ring 122 and a tray 121 and fixing the support ring 122 and the tray 121 with a M10 (JIS) hexagonal bolt 126 and hexagonal nut 126 and washer 125.

The apparatus was set operating as aimed at two months' continuous operation. From the start of the operation, the pressure in the lower part of the distillation column was

observed to rise with the elapse of time. Thus, the apparatus was stopped after one month's operation. When the interior thereof was visually inspected, the adhesion of a polymer to the wall of the column and the tray-fixing bolt was observed and the accumulation of polymer on the outer peripheral parts of the trays was also observed. The amount of the polymer so accumulated was about 10 kg per stage.

#### Conclusion of the Experiments

Experiment is mainly performed in accordance with Experiment 1. In Experiment 2, a vertical clamp was used in fixing the trays to the support rings. In Comparative Experiment 1, a horizontal claim was used in fixing the trays to the support rings. In Comparative Experiment 2, a liquid passing opening on the welding line between the support ring and the wall of the column is eliminated and the trays are fixed as illustrated in Fig. 11.

In Experiment 2, the apparatus was continuously operated for two months and then brought to stop. At that time, the amounts of the polymer so accumulated was about 3 kg per stage.

In contrast, in Comparative Experiments 1 and 2, the apparatus was stopped after one month's operation, since the pressure in the lower part of the distillation column was observed to rise with the elapse of time. At that time, the amounts of the polymer so accumulates were about 50 kg per stage for the former and about 10 kg per stage for the latter.

As described above, in accordance with the present invention, the apparatus can be operated longer than, more than 2 times, those using the horizontal clamp and without liquid passing openings.

III. The undersigned declares that all statements made

herein are true; and further that these statements were made  
with the knowledge that willful false statements and the like  
so made are punishable by fines or imprisonment, or both under  
Section 1001 of Title 18 of the United States Code and that  
5 such willful false statements may jeopardize the validity  
of the application or any patent issuing thereon.

10      May 17, 2005  
Date

Tetsuji Mitsumoto  
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